ENVIRONMENTALLY BENIGN SILICON SOLAR CELL MANUFACTURING

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ABSTRACT: The manufacturing of silicon devices - from polysilicon production, crystal growth, ingot slicing, wafer cleaning, device processing, to encapsulation - requires many steps that are energy intensive and use large amounts of water and toxic chemicals. In the past two years, the silicon integrated-circuits (IC) industry has initiated several programs to promote environmentally benign manufacturing, i.e., *manufacturing practices that recover, recycle, and reuse materials resources with a minimal consumption of energy.* Crystalline-silicon solar photovoltaic (PV) modules, which accounted for 87% of the worldwide module shipments in 1997, are large-area devices with many manufacturing steps similar to those used in the IC industry. Obviously, there are significant opportunities for the PV industry to implement more environmentally benign manufacturing approaches. Such approaches often have the potential for significant cost reduction by reducing the energy use and/or the purchase volume of new chemicals and by cutting the amount of used chemicals that must be discarded. This paper will review recent accomplishments of the IC industry initiatives and discuss new processes for environmentally benign silicon solar-cell manufacturing.
Keywords: c-Si - 1: Manufacturing and Processing - 2: Environmental Effect - 3

1. INTRODUCTION

With the worldwide photovoltaic (PV) solar energy market expanding rapidly and the demand outpacing supply, the crystalline-silicon solar PV module is finally becoming a commercially viable product. Most of the newcomers and the capacity expansions of existing producers are based on either single-crystal silicon grown by the Czochralski (Cz) method or polycrystalline silicon by casting. There are also increasing number of suppliers who are developing production equipment for the industry based on best-known practices (BKPs) of silicon solar cell and module processing. Because solar electricity generation is a large-area application, it is likely the PV industry will eventually use more silicon than even the integrated-circuits (IC) industry. It is important at this stage to review the environmental impact of the rapidly growing silicon PV industry and to find opportunities for improving the energy efficiency and productivity and reducing environmental impact.

The manufacturing of semiconductor silicon devices - from polysilicon production, crystal growth, ingot slicing, wafer cleaning, device processing, to encapsulation - requires many steps that are energy intensive and use large amounts of water and toxic chemicals. In the past two years, the IC industry has initiated several programs to promote environmentally benign manufacturing, i.e., manufacturing practices that recover, recycle, and reuse materials resources with a minimal consumption of energy. One of the programs is the establishment of the *Engineering Research Center for Environmentally Benign Semiconductor Manufacturing* on April 15, 1996, with an initial funding of US$10 million from the United States National Science Foundation (NSF) and Semiconductor Research Corporation (SRC). Crystalline-silicon modules accounted for 87% of the worldwide PV module shipments in 1997 and 93% of those modules shipped for outdoor applications [1]. Silicon PV module manufacturing have many steps similar to those used in the IC industry. Although the annual sales of the worldwide silicon PV module industry is about 400 times smaller than that of the IC industry, the PV industry consumes about 10% of the worldwide polysilicon production. Obviously, there are significant opportunities for the silicon PV industry to learn from new developments in the IC industry and to implement more environmentally benign manufacturing approaches. Such approaches often have the potential for significant cost reduction by reducing the purchase volume of new chemicals and by cutting the amount of used chemicals that must be discarded. Because PV manufacturing has lower semiconductor material-quality requirements than IC manufacturing, some lower cost and more environmentally sound processes may be acceptable for PV manufacturing even though they don't meet the stricter requirements of IC manufacturing.

It is not possible to review here in detail all the steps involved in the manufacturing of silicon PV modules. We will simply highlight some areas where we think opportunities exist to make the module manufacturing process more environmentally benign.

2. INDUSTRY INITIATIVES AND RESOURCES

Three recent initiatives by the semiconductor industry in the environmentally benign manufacturing area could produce results that also benefit the PV industry: (1) The National Science Foundation and the Semiconductor Research Corporation jointly established the *NSF-SRC Engineering Research Center for Environmentally Benign Semiconductor Manufacturing* on April 15, 1996 [2]. SRC is a consortium of 65 corporations and government agencies that plans, directs, and funds the semiconductor industry's pre-competitive, long-term research [3]. (2) In April 1997, the international trade association for semiconductor industry, Semiconductor Equipment and Materials International (SEMI), created a new *Environmental Health and Safety Division* that will explore worldwide environmental priorities for the industry [4]. (3) In October 1997, the Electric Power Research Institute (EPRI) and the SEMATECH Corp. formed a new center, the *EPRI Center for Electronics Manufacturing*, to address productivity, environmental, and energy issues in the electronics industry [5]. SEMATECH is a non-profit R&D consortium of U.S. semiconductor manufacturers.

The NSF-SRC Center carries out research in six areas of semiconductor manufacturing: water conservation, plasma processes, wet chemicals, chemical-mechanical polishing, emission of organics, and risk-assessment studies. Some of these research results are presented in a weekly teleconference seminar series hosted by the four participating universities: University of Arizona, Massachusetts Institute of Technology, Stanford University, and University of California-Berkeley [6]. A good source of information on the Environmental, Safety, and Health (ES&H) goals of the semiconductor industry is the ES&H Section of the United States National Roadmap for Semiconductors [7]. In addition to these semiconductor industry organizations, the United States National Photovoltaic Environmental, Health and Safety Information Center [8] regularly publishes information on PV ES&H-related issues [9].

3. POLYSILICON PRODUCTION

For the feedstock material for crystal growth, the silicon PV industry has been relying on rejected materials from the IC industry. These rejected materials, about 2,100 metric tons in 1997, amount to about 10% of the semiconductor-grade polysilicon used by the IC industry. This arrangement worked well until 1995 when shortage of polysilicon feedstock began to drive up cost and limit the growth of the silicon PV industry. If the PV industry continues to grow at the present rate, which in recent years has been higher than the growth rate of the IC industry, and if crystalline silicon continues to be the dominant technology of the PV industry, then we must develop new sources of solar-grade polysilicon. There are two possibilities: (1) build new factories dedicated to the production of low-cost (< US$10/kg), solar-grade polysilicon, and (2) find new ways to use the rejected silicon that is not currently being used, for example, purifying the about 30% of silicon lost from wafer-cutting operations (kerf loss) of semiconductor-grade polysilicon into solar-grade polysilicon. The purity requirements for solar-grade polysilicon, according to the Solar-Grade Silicon Stakeholders Group, are the following: it is preferred that polysilicon have either B or P doping, with no compensation; resistivity at $25^{O}C$ should be greater than 1 ohm-cm; oxygen and carbon should not exceed the saturation limits in the melt; and the total non-dopant impurity concentration should be less than 1 ppma [10].

More than 98% of semiconductor-grade polysilicon is produced by the trichlorosilane ($SiHCl_3$) distillation and reduction method [11,12]. The trichlorosilane is manufactured by fluidizing a bed of fine pulverized metallurgical-grade silicon (MG-Si), which is more than 98% silicon, with hydrogen chloride in the presence of a copper-containing catalyst. The MG-Si, which costs about US$1/kg, is produced by the reduction of natural quartzite (silica) with coke (carbon) in an electric arc furnace. This method of polysilicon production is very energy intensive [13], and it produces large amounts of wastes, including a mix of environmentally damaging chlorinated compounds. About 80% of the initial metallurgical-grade silicon material is wasted during the process. In addition, the semiconductor-grade polysilicon material produced by this method far exceeds the purity requirement of the PV industry, and the cost (over US$50/kg, with most of it attributable to the $SiHCl_3$ processes) is several times higher than what the PV industry can afford [14]. Every watt of crystalline silicon PV module generating capacity requires roughly 20 g of polysilicon. Thus, if the cost of solar-grade polysilicon is US$20/kg, the cost of polysilicon per watt of a crystalline-silicon PV module is US$0.40. It is obvious that less complicated, less energy intensive, more efficient, and more environmentally benign methods need to be developed to meet the cost and quality requirements of the PV industry. New methods of producing solar-grade polysilicon should either be chlorine free or recycle chlorine internally to reduce cost and avoid damage to the environment.

3.1 Low-Temperature, Chlorine-free Processes for Polysilicon Feedstock Production

The National Renewable Energy Laboratory (NREL) and Sandia National Laboratories (SNL), with funding from the Initiative for Proliferation Prevention (IPP) Program, has initiated a joint research program with the Intersolarcenter to study new chlorine-free methods of producing solar-grade polysilicon. So far, the most promising method developed by this project is one that uses MG-Si and absolute alcohol as the starting materials. This new process requires only 15 to 30 kWh of energy per kg of polysilicon produced vs. about 250 kWh/kg of the trichlorosilane method. The silicon yield (polysilicon and the main by-product, silica sol) is in the 80% to 95% range vs. 6% to 20% for the trichlorosilane method. The eventual cost goal is US$10 per kg of solar-grade polysilicon.

The basic processing stages of this chlorine-free polysilicon production process are the following:

1. The reaction of metallurgical-grade silicon with alcohol proceeds at 280°C in the presence of a catalyst:

$$Si + 3\ C_2H_5OH \xrightarrow{catalyst} Si(OC_2H_5)_3H + H_2\ . \quad (1)$$

2. The disproportion (i.e., simultaneous oxidation and reduction) of triethoxysilane in the presence of a catalyst will lead to the production of silane and tetraethoxysilane:

$$4\ Si(OC_2H_5)_3H \xrightarrow{catalyst} SiH_4 + 3\ Si(OC_2H_5)_4\ . \quad (2)$$

3. Dry ethanol and such secondary products as high-purity SiO_2 or silica sol can be extracted by hydrolysis of tetraethoxysilane. The alcohol will be returned to Stage 1.

$$Si(OC_2H_5)_4 + 2\ H_2O \text{ ----- } SiO_2 + 4\ C_2H_5OH\ . \quad (3)$$

4. Silane is decomposed pyrolytically to pure silicon and hydrogen at a temperature of about 900°C:

$$SiH_4 \xrightarrow{850^\circ - 900^\circ C} Si + 2\ H_2\ . \quad (4)$$

The purity requirements for solar-grade silicon are not as high as those for electronic applications. Thus, the silane will undergo a simplified cycle of purification, and at Stage 4 the less expensive and less energy-consuming process of a fluidized bed reactor can be used, instead of the well-known Siemens Process [11].

3.2 Purification of Metallurgical Silicon

NREL and ENEA (National Agency for New Technologies Energy & Environment) have proposed a novel method of producing solar-grade polysilicon by directly purifying MG-Si pellets. The process uses the very large surface areas, produced by porous silicon etch on the surfaces of the silicon wafer, as sites for gettering impurities in the subsequent high-temperature annealing. The details of this process will be presented seperately at this conference [15].

3.3 New Sources of Silicon Waste from the Electronic Industry

When wafers are sliced from silicon ingots using a multiple-wire saw, a layer of silicon about 250 micrometers thick is lost per wafer. This kerf loss is higher for inner-diameter (ID) saws. Depending on the wafer thickness, this kerf loss represents from 25% to 50% of the ingot material, several times the quantity of the material that is presently used by the PV industry. Presently, the solar industry uses mainly top and tails, pot scrapes, and rejected wafers from the IC industry [14]. If a method can be developed to produce solar-grade polysilicon by purifying the kerf remains of semiconductor-grade ingots, enough polysilicon would be generated for over 300 MW/year of crystalline-silicon solar cells, i.e., more than two times the size of the current silicon solar-cell production.

4. CRYSTAL GROWTH

There are four types of crystalline-silicon solar cells: single-crystal, polycrystalline, ribbon, and silicon film deposited on low-cost substrates. In 1997, market shares of the worldwide PV cell and module shipment for the four types of crystalline-silicon solar cells were 49.6% for single-crystal, 34.0% for polycrystalline, 3.2% for ribbon, and 0.4% for silicon film [1]. Crystal growth from silicon melt generates relatively few wastes. The main concern is the energy required and the amount of argon gas used during crystal growth. Electricity and argon needed for Cz growth are the highest among the four types of silicon materials [13]. Recently, however, the world's largest manufacturer of Cz silicon solar cells, Siemens Solar, Industries, announced a joint project with the Northwest Energy Efficiency Alliance to cut the amount of electricity used to grow crystals and yield savings of 40% to 50% [16].

5. WAFER SLICING AND CLEANING

5.1 Wafer Slicing

In the last six years or so, the PV industry has made the transition from using ID saws for wafer slicing to using multiple-wire saws. Multiple-wire saws can improve wafer yield per unit length of ingots by over 50% because of lower kerf loss and thinner wafers. However, wafer slicing is still one of the most expensive processes in silicon solar-cell manufacturing because of the large quantities of consumables (stainless-steel wires and abrasive slurry) and the kerf loss. During wafer slicing, ingots are bonded to a ceramic submount with hot-melt adhesive and sliced into wafers with multiple steel wires to which an abrasive slurry is fed. The slurry is composed of silicon carbide (SiC) and mineral-oil-base or glycol-base slurry vehicle. Oil-base slurry is commonly used by the PV industry. Compared to the water-soluble, glycol-base slurries more commonly used by the IC industry, oil-base slurry produces more environmentally damaging wastes and requires more extensive wafer cleaning. The added cost and the process changes needed for the PV industry to switch over to glycol-base slurry need to be investigated. Methods of the proper disposal or recycling of the stainless-steel cutting wires also need be studied, as does the effective recovery of the SiC in the slurry. The development of water-base slurries will also help reduce cost and environmental damage.

5.2 Wafer Cleaning and Etching

The cost of chemical waste disposal is high. It is important for the PV industry to find ways to reduce chemical consumption and waste generation through *source reduction, recovery, recycle, reuse, and substitution*. Because wafer cleanliness for PV is not as critical as for IC manufacturing, a safe choice, in terms of making sure highest quality and most extensive cleaning procedures are used, is not necessarily the right choice in terms of cost reduction and environmental safety. Certain methods, such as dry cleaning processes, although not adequate for the IC industry standards, may be sufficient for the PV industry. For example, centrifugal shear carbon dioxide cleaning [17] is worth considering as an alternative to organic solvent and/or hot detergent cleaning methods for wafer degreasing and cleaning after slicing. This process uses carbon dioxide in three coexisting phases: liquid, supercritical, and dense-gas. Operating temperature ranges from 298 K to 310 K, and pressure ranges from 56 ATM to 100 ATM. CO_2 gas is non-flammable, non-combustible, and non-corrosive, and is abundant, inexpensive, and reusable. Compressing CO_2 at about 70 ATM and at temperatures below its critical temperature (305 K) liquefies the gas. Compressing CO_2 above its critical temperature and pressure (72.9 ATM) does not cause a phase change, yet the density of the gas may be liquid-like. Static and dynamic cleaning processes employing the multi-phase CO_2 system have been developed. The excellent cleaning abilities derive from a combination of solvent cleaning power and, in the dynamic processes, by physical cleaning action. Although CO_2 at these pressure and temperature conditions presents densities comparable to other cleaning agents (about 500 g/L), it has a viscosity comparable to gases. For instance, it is from 10 to 30 times smaller than 1,1,1-trichloroethane (TCA).

For texture etching and/or surface damage removal, most of the PV industry has been using sodium hydroxide etchant. NaOH etchant is considerably cheaper and easier to dispose of than the conventional hydrofluoric-nitric-acetic acid etchant. In an NREL-funded program, Siemens Solar Industries found that the caustic waste per wafer generated by the saw-damage-removal etching process was reduced by about 20% after the switch from ID saws for wafer slicing to multiple-wire saws [18]. This is because the wire-sawn wafers require less etching to remove saw damage.

The Photovoltaic Device Fabrication Laboratory (PDFL) at Sandia National Laboratories has had a program to continuously monitor and reduce chemical usage since the laboratory began operations in 1989. The chemical waste generation has been reduced by nearly 75% since 1990 by using three procedures: (1) eliminate and/or replace hazardous chemicals, (2) recycle chemicals, and (3) reduce usage of remaining hazardous chemicals. A key feature of this effort was use of statistical experimental designs to screen chemicals for their effectiveness in wafer cleaning and statistical process control to ensure that chemical-reduction changes did not impact the manufacturing process. An experiment was performed that used statistical designs to examine the effect of 22 different parameters associated with chemical cleaning of wafers. The experiment was able to eliminate a popular, but expensive, chemical (hydrogen peroxide) that is widely used in the IC industry for cleaning silicon wafers. Subsequent experiments and changes in procedure have eliminated sulfuric and phosphoric acids from PDFL.

Hydrofluoric (HF) acid solution is used for wafer cleaning, dopant oxide removal, and diffusion tube and quartz cleaning. It accounts for a very large percentage of the total hazardous waste generated by silicon solar-cell manufacturing. It is possible to reprocess used HF solutions using reverse osmosis [19]. A cost-benefit analysis indicates that, for a system with a capacity of 1000 gal/day, about one US dollar is the net savings for every gal of HF solution reprocessed [20].

5.3 Optimizing Water Use and Reuse

The semiconductor industry worldwide spends as much on ultrapure water as on wet chemicals for wafer processing, about US$700 million each in 1996 [21]. The net-feed water use by the IC industry averages about 30 gallons per square inch of wafer processed in 1997. The current United States National Roadmap for Semiconductors recommends decreasing the net-feed water use to 10 gallons/in^2 in 2000 and 2 gallons/in^2 in 2012 [22]. The NSF/SRC Center is studying methods to decrease water usage by more efficient rinse processes, water conservation in cooling, scrubbing and washing, and by lowering idle flows [23].

The silicon PV industry, of course, uses significantly less water per wafer than the IC industry. However, the value of the final product per wafer of the PV industry is orders of magnitude less than that of the IC industry. Obviously, water use by the PV industry is a significant cost factor that needs to be carefully studied. It is also not clear whether the PV industry really needs to use the same high-purity deionized water as that used by the IC industry. Water purity in the semiconductor process is typically measured in a bath with a resistivity monitor. The resistivity is inversely proportional to the ionic concentration of chemicals. At 25^{o}C, water can exhibit a resistivity of 18.2 megaohm-cm if no impurities are present. The American Society for Testing and Materials has set four types of requirements for electronic water [24]. The resistivity (in megaohm-cm) at 25^{o}C for the highest-grade electronic water, Type E-1, should be above 18.0 for 95% of the time and never less than 17.0. Type E-2 should be above 17.5 for 90% of the time and not less than 16.0. Type E-3 should be above 12.0. Type E-4 should be above 0.5. Type E-1 water, which costs 1 to 2 US dollars per 100 gallon, is intended for use in the production of devices having line widths below 1.0 micrometer. Type E-2 water is for line widths below 5 micrometers and is probably more than adequate for even the most critical wafer-cleaning needs by the PV industry. Even Type E-3 may be adequate in most cases.

Reclaiming water is also an important issue to be investigated by the PV industry. For every dollar spent to generate and process ultra-purity water by the IC industry, about $0.60 is spent treating the industrial wastewater. According to SEMATECH, 70% of its members are reclaiming at least some of their water. Careful design of the methods of recycling used water back to the feed makeup for the water purification plant and the methods of reclaiming water for use in support processes, such as cooling and gas scrubbing, is important for reducing the cost of PV manufacturing.

6. SOLAR CELL PROCESSING

For junction diffusion, either a belt furnace or a tube furnace is typically used. Tube furnaces traditionally use a $POCl_3$ liquid source dopant, which generates toxic P_2O_5 and Cl_2 effluents and requires frequent cleaning of diffusion tubes using HF solutions. Belt furnaces are more environmentally benign because they can use water-soluble, non-toxic, spin-on or spray-on dopants or vapor dopants and do not require HF cleaning. Optically enhanced doping methods, such as the solar furnace doping proposed by NREL [25], are also environmentally benign alternatives.

Edge trimming to remove electrical shorts between the front and back junction can be done either by laser cutting or plasma etching. Because of its very high throughput, $CF_4 + O_2$ plasma etching is commonly used by the PV industry. However, CF_4 is one of the perfluorinated compounds (PFCs) that has no known natural destruction mechanisms, and thus, has a large global-warming potential [26]. The PV industry needs to either find an alternative to the CF_4 and O_2 plasma etch or improve the effluent treatment to include PFC capture and recycling.

Antireflection coatings can be deposited by vacuum evaporation, plasma deposition, atmospheric-pressure chemical vapor deposition, and spin-on liquids. Silane, which is pyrophoric, is commonly used in depositing silicon nitride antireflection coatings [27]. A safer alternative, chlorosilane, which is non-pyrophoric, has been used successfully by the IC industry for silicon nitride depositions [9].

For metal electrodes on the solar cells, it is not desirable to use silver-tin-lead solder baths after screen printing to enhance the conductivity of the metal grids because of the added lead content to the cells.

7. MODULE ASSEMBLY

7.1 No-Clean Flux

Flux, typically a derivative of pine resin, is applied to cell interconnection strips before soldering to act as a deoxidizer and to ensure better adhesion between the solder and solar cells. Conventional flux leaves residues on the cell surfaces that need to be cleaned with a chlorofluorocarbon (CFC) compound. CFCs are known to cause ozone depletion in the atmosphere [28]. Recently, water-soluble fluxes and no-clean fluxes, low-residue fluxes that could be left on the solar cell after soldering have become widely available [29]. In an NREL-funded program, Siemens Solar was able to eliminate the CFC usage in the manufacturing facility by switching from conventional solder paste to a “no-clean” solder paste [18]. The no-clean process both eliminates the environmental damaging CFC emissions and reduces costs. Siemens Solar also found that, by using a water-soluble flux, the CFC usage can be reduced by about 60% over a conventional flux. However, it appeared that water rinse of the cells retained moisture during the lamination sequence and caused module reliability problems.

7.2 Lead-Free Solder

Lead is a well-known hazard to human health. When disposed of in landfills, it can leach into soils and pollute ground water. It is important for the PV industry to remove or minimize the use of lead in modules so that proper disposal at the end of module life will not become a problem [30]. For example, some European countries have proposed a ban on the landfill disposal of electronic products containing lead. There are two sources of lead in a crystalline-silicon PV module: solder-dipped electrodes and solder-coated copper ribbons. The practice of dipping-coating solar-cell contact electrodes is no longer necessary with modern screen-printed electrodes, but it is still used by a significant number of module manufacturers. The tin-lead solder that coats the surfaces of copper ribbons for tabbing strips is needed to prevent the oxidation of copper and to improve the solderability of the ribbons. However, lead-free alternatives to tin-lead solder have been investigated extensively by the printed-circuit-board industry [31]. For example, the National Center for Manufacturing Sciences in Ann Arbor, Michigan, has recently completed a US$10-million project that evaluated 79 lead-free solder alloys and found seven promising replacement candidates [32]. The International Tin Research Institute (ITRI) in Middlesex, England, has also done extensive studies on lead-free solder alloys [33].

A very promising alternative for the tin-36% lead-2% silver ribbon coatings commonly used by the PV industry is the tin-3.5% silver alloy. It is identified as a promising alternative for the standard tin-37% lead alloy by both NCMS and ITRI. The silver in the alloy is needed to increase the pull strength of the ribbon. The $221^{o}C$ melting temperature, although higher than the $183^{o}C$ for the standard tin-lead alloy, is acceptable. Because it is a binary alloy, it should have excellent stability. Its resistance to high-temperature fatigue is also good. The present cost of the alloy, at US$0.10 per cubic centimeter, is about twice the standard tin-lead alloy, which is the cheapest of the tin alloys. The cost difference between the tin-silver alloy and the tin-lead-silver alloy is small.

8. CONCLUSIONS

As the silicon PV industry continues to rapidly expand, the environmental impact of its manufacturing processes and products will receive increasing attention. It is particularly important for a renewable energy technology to address its environmental impact during manufacturing because one of the primary benefits of renewable energy generation is its low environmental impact. We have discussed several alternative approaches in this paper that are both cost effective and environmentally benign. However, the manufacturability and reliability of most of these alternative approaches need further investigation. We propose that the silicon PV industry form an association of government laboratories, equipment suppliers, and cell and module manufacturers to promote more environmentally benign manufacturing approaches. This association can also coordinate PV industry’s interactions with the environmental associations of the integrated-circuits and printed-circuit-board industries mentioned in this paper.

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